Referee #2

GENERAL REMARKS

The manuscript reports results from a multi-annual study on aerosol optical properties and scavenging effects, observed at the Puijo measurement station in Kuopio, Finland. Observed properties include number concentrations of different aerosol modes, size distributions, scattering and absorption coefficients. The authors investigate the impact of environmental parameters (temperature, relative humidity, in-cloud and clear sky conditions) on the scavenging and wet deposition of various aerosol parameters. The experimental part of the study is very well designed and carefully conducted. The resulting data are of high quality and of high relevance for the investigation of the aerosol indirect effect because the data set covers a long period in time and thus a large variety of weather situations and atmospheric conditions. The interpretation of the presented data, however, remains largely on the level of describing observed phenomena, whereas modelling studies for quantitative analyses are lacking. In summary, the topic of the manuscript fits well into the scope of the journal. The manuscript can be accepted for publication in ACP after major revisions have been considered which are specified in the following.

> We thank Referee #2 for carefully reading the manuscript and providing valuable suggestions to improve the manuscript. Our point-by-point replies to the specific comments are given below. The referee comments are highlighted in blue with our responses in black and indented. The excerpts from the revised manuscript are in Italics and the font size is smaller.

SPECIFIC COMMENTS

1. The authors discuss the influence of environmental parameters like temperature and relative humidity on the properties of the sampled aerosol, and in particular of the scavenging efficiency. In the abstract and also later in the main text of the manuscript, the authors discuss the impact of air temperature on the observations just as if there is a direct dependency of temperature on the observed properties. However, particularly the impact of air temperature on the observed effects is only of indirect nature, since the aerosol population and constituents change with season and thus with temperature because of changing sources. An even better wording could be to describe the link between temperature and the observed effects as a correlation instead of a dependency. This fact should be clearly stated because in the current manuscript it reads like there is a clear temperature-dependence on aerosol properties like number concentrations etc.; see e.g. lines 19 to 22 of the abstract. The same is probably true for the impact of relative humidity since scavenging efficiencies at the same relative humidity level may change between, e.g., spring and fall conditions with different aerosol chemical compositions. Again, the effect of relative humidity on the hygroscopic behaviour of the aerosols is not only related to the level of relative humidity but also to the difference in chemical composition.

> We thank the referee for this very good comment and will revise the manuscript accordingly in order to emphasize the indirect nature of the environmental parameters, such as temperature, on the studied aerosol properties. We chose temperature as the main environmental parameter because it is, despite of its indirect nature, the easiest parameter to be used in the analysis and in comparison with model calculations. Furthermore, similar representation has been used in several previous studies, which enabled us to carry out straightforward comparison with the literature.

2. In its current version, the analyses presented in Figures 3 to 6 may suffer from pooling different aerosol chemical compositions and thus different hygroscopic growth behaviour into single bins for temperature and relative humidity. The authors describe this effect on page 6 lines 12 to 25 for the data shown in Figure 4, but not in a quantitative manner. The missing quantification however, makes the data

less valuable for modelling studies since key properties are missing in the analysis. To overcome this limitation, it might be worthwhile to investigate, e.g., the contributions of various parameters on the variability of the fraction of scavenged absorbing material at -7◦C (Figure5b). In the current analysis, this fraction is centred at 0.45 with a P10 value below 0.1and a P90 value close to 0.8. A similar exercise could be conducted for most of the other analyses.

We agree with the referee that pooling too much data causes variation in the analysis results and that more detailed information would increase the value of the presented data. Unfortunately, we do not have, e.g., long-term mass spectrometry or updraft measurements at the cloud base which would help us in investigating the effect of aerosol chemistry and mixing state on the observed D50 values in more detail. However, we consider that providing average BC scavenging efficiencies and connecting it with cloud dynamics through the calculated D50 values provides a valuable data set for model evaluation purposes when low altitude clouds are considered.

3. In the introduction section (page 2, lines 13 to 25), the authors describe the interaction of aerosol particles with water vapour. This section requires rewriting for several reasons. The effect of hygroscopic growth at relative humidity < 100%, and even more important, the effect of cloud condensation nucleus activation is not related to microphysical processing but to water uptake by hygroscopic material. A more precise description is needed here. Later in this paragraph, the authors discuss that due to the different hygroscopic properties which favour scavenging of water-soluble light-scattering material, light absorbing aerosol is enriched in cloud-processed air parcels compared to its initial state, the cooling effect of liquid-water clouds is reduced com-pared to the warming effect of light absorbing material. A more detailed description and references are needed here.

We will rephrase the paragraph in order to state more clearly that black carbon (BC) resides typically in relatively small particles, which activate unlikely to cloud droplets and thus are also less likely to be wet scavenged, whereas the more hygroscopic fraction is typically found in larger particles and is more likely scavenged through cloud processes. This holds at least in low altitude clouds where below cloud washout is relatively less important than in cloud scavenging. Thus, the lifetime of BC is expected to be longer.

4. In Figure 5, the authors present a regression analysis of temperature dependence of the fractions of lights scattering and light absorbing material; see Section 3.2.2.The results of this regression analysis are also listed in the main conclusions of the manuscript. The authors explained that the errors of observations were taken into account by performing a Deming regression analysis. The applied method is a suitable choice, but the statistical significance of the obtained results needs to be discussed.

We introduced the confidence levels and added a short statement about the significance:

The slopes for scavenging efficiencies were calculated to be approximately 0.0052 (95% CL for slope: 0.0042-0.0062) for scattering (Fig. 5a) and 0.0034 (0.0022-0.0046) for absorption (Fig. 5b). Which means that the effects are statistically significant and corresponding to approximately 5 percentage unit change for scattering and 3 for the absorbing material in the scavenged fraction at 10°C change in temperature, respectively.

MINOR ISSUES

Page 2, line 39: The authors state that the effect of clouds and precipitation on aerosol properties has been studied in a few campaigns. However, there have been many field campaigns conducted on this topic, which is also reflected in the list of references given in the manuscript. An adequate restatement is requested.

We reformulated the sentence which currently reads as:

The effect of clouds or precipitation on aerosol optical properties has also been studied in several campaigns (Zhang et al., 2012; Berkowitz et al., 2011; Hyvärinen et al., 2011; Chaubey et al., 2010; Marcq et al., 2010; Yamagata et al., 2009; Cozic et al, 2007; Latha et al., 2005) and by conducting model calculations (e.g., Browse et al., 2012; Croft et al., 2009).

Page 5, line 6: The minimum diameter for the total aerosol should be stated here.

We added the minimum diameter which is the same for both measurement lines.

The total number concentration (N_{tot}) is defined as the number concentration of all particles ranged from 3 or 7 nm to 800 nm (see section 2.2).

Page 6, line 35: At some positions in the manuscript an article is missing, e.g., “First, sampling system with...” should read “First, a sampling system with...”. Checking the manuscript text is recommended.

We will check the grammar throughout the revised manuscript as suggested.

Page 7, line 12: a comma should be added after “Third”.

The sentence has been rephrased slightly for clarification:

The third and last possible source of dispersion resides in long term measurements.